ELSEVIER

Surface Science 499 (2002) L141–L147

SURFACE SCIENCE

www.elsevier.com/locate/susc

Surface Science Letters

5f Resonant photoemission from plutonium

J. Terry [a,1], R.K. Schulze [a], J.D. Farr [a], T. Zocco [a], K. Heinzelman [b], E. Rotenberg [b], D.K. Shuh [b], G. Van der Laan [c], D.A. Arena [d], J.G. Tobin [d,*]

[a] *Los Alamos National Laboratory, Los Alamos NM 87545, USA*
[b] *Lawrence Berkeley National Laboratory, Berkeley CA 94720, USA*
[c] *Daresbury Laboratory, Warrington WA4 4AD, UK*
[d] *Lawrence Livermore National Laboratory, Livermore CA 94550, USA*

Received 19 September 2001; accepted for publication 29 October 2001

Abstract

Experimental resonant photoemission (ResPes) results for α-Pu and δ-Pu bulk samples are presented and compared to the results of an atomic model calculation. Both Pu samples exhibit limited agreement with the atomic model calculations. As expected, α-Pu appears to have more 5f valence band delocalization than δ-Pu. Evidence of an enhanced sensitivity to surface corruption, by using synchrotron radiation as the excitation, is presented.

Keywords: Polycrystalline surfaces; Surface structure, morphology, roughness, and topography; Synchrotron radiation photoelectron spectroscopy

The valence electronic structures of the actinide metals and alloys in general and plutonium (Pu) in particular remain mired in controversy [1–5]. Interestingly, the various phases of Pu metal provide a microcosm of the metallic actinides as a whole [6–9]. Thus, unravelling the nuances of the interplay of electronic and geometric structures in Pu will illuminate the properties of all transuranic metals. In a sense, the behavior of the Pu 5f electrons is completely counter-intuitive. The dense phase, α, has some semblance of delocalization in the 5f valence bands and can be treated theoretically within single electron models such as the local density approximation (LDA). The α phase is monoclinic, which is a low symmetry ordering [10]. The less dense δ-phase is fcc and exhibits evidence of localized and/or correlated electronic behavior. The fcc is a high symmetry phase which is normally associated with superior wave function overlap in d state metals. But herein is the key: the linear combinations of the 5f's do not produce the nicely lobed wave functions with symmetry about the *x*, *y*, *z* and diagonal axes, as occurs for d states. Instead, the linear combinations of f-states have oddly lobed and badly directed wave functions

* Corresponding author. Tel.: +1-925-422-7247; fax: +1-925-423-7040.
E-mail address: tobin1@llnl.gov (J.G. Tobin).
[1] Present Address: Ill. Inst. of Tech. and the Adv. Ph. Source, ANL, Chicago, IL, USA.

0039-6028/01/$ - see front matter
PII: S0039-6028(01)01757-5